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Thermoelectric power of graphite

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A complete record of the thermoelectric power (t.e.p.) of graphite and its variation with temperature as also a theory to account for such a variation have long been wanting since none of the early experiments (Gottstein 1914, Pirani & Fehse 1929, Fukuda & Saito 1938) could provide any consistent data on the subject. The first methodical investigation of the t.e.p. of polycrystalline graphite as a function of temperature was undertaken by Tyler & Wilson (1953) in the range 20°K–280°K. They found the t.e.p. to be negative over the entire temperature range with a minimum between 30°K and 40°K. But they were unable to interpret the data. Loebner (1956) studied the dependence of the t.e.p. of carbons on the temperature of heat treatment but could provide no suitable explanation for graphitised specimens. But still interesting problems like finding out the effect of phonon-drag on the t.e.p. or making an idea about the carrier mobilities,

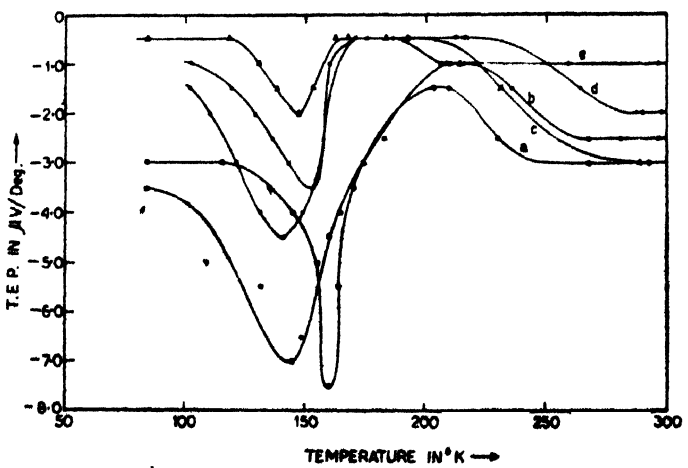

Fig. 1. Temperature variation of thermoelectric power of graphite.
a = crystal 2(a) ... temperature gradient along basal plane
b = crystal 3(a) ... -do-
c = crystal X_1 ... -do-
d = crystal 1(c) ... temperature gradient along c-axis
e = compacted natural flakes 2(c) -do-

Fermi energy etc. could not be touched upon since no dependable single crystal data were available. Blackman, Dundas & Ubbelhode (1960), however, undertook the measurements of principal t.e.p's with pyrolytic graphite crystals, natural single crystals and compacted natural flakes. They observed that for the *a*-axis direction t.e.p. were negative except for three pyrolytic samples while about the *c*-axis all the samples including compacted natural flakes (no single crystal was used for *c*-axis measurements) showed a positive t.e.p. They inferred that, along the *c*-direction holes and along *a*-axis electrons are predominant current carriers. Such a situation is evidently rather difficult to release vis-a-vis the electronic picture of graphite. Further, these authors did not report any temperature variation of t.e.p. with natural single crystals expect with one crystal compact for *c*-axis direction only. We have, therefore, undertaken to study both the principal t.e.p.'s of graphite against copper, following the method of Loebner (1956), with some well-developed single crystals of Ceylon graphite over the temperature range 84°K–300°K.

From these observations it is found that t.e.p. is negative in both *a*-axis and *c*-axis directions and that their values at room temperature are practically of the same order of magnitude (2–3 $\mu v/°C$). Even with a compact made of natural flakes the *c*-axis t.e.p. is negative. These observations are thus not in agreement with those of Blackman *et al* (1960). Temperature variation of t.e.p.'s of different samples of graphite for different directions (*a*- and *c*-axis) are shown in figure 1. The values of t.e.p. remain negative throughout the entire temperature range. There is, however, a singularity in the curves between 140°K and 160°K.

Assuming an overlap model for graphite with equal number of electrons and holes and considering phonon-drag to be negligible in the temperature range 84°K–300°K thermoelectric power (S) may be expressed by the relation (Klein 1964),

$$S = \frac{k}{e}\,\frac{1-b}{1+b}\left[\frac{2F'(\zeta/kT)}{F_0(\zeta/kT)} - \frac{\zeta}{kT}\right], \qquad \ldots (1)$$

where b is the ratio of mobilities of electrons and holes, ζ the Fermi energy, $F_0(\zeta/kT)$ and $F'(\zeta/kT)$ are the Fermi-Dirac functions whose values are obtained from the table by McDougall & Stoner (1938) and the rest of the symbols have their usual significances. With the help of this relation and with the experimental values of S at different temperatures, b for different samples of graphite have been calculated, assuming $\zeta = 0{\cdot}022$ ev. as obtained by McClure (1957) from a theoretical analysis of the DeHaas-Van Alphen data of graphite on the assumption of an overlap model. Calculations have also been made with $\zeta = 0{\cdot}005$ ev. as suggested by Klein (1964). The results are diagrammatically represented in figure 2 where the temperature variation of b for the crystal X,

obtained from Hall effect and conductivity measurements are also included for comparison. It is observed that in the curves obtained with $\zeta = 0{\cdot}022$ ev. the singularity in the t.e.p. temperature curves is better represented.

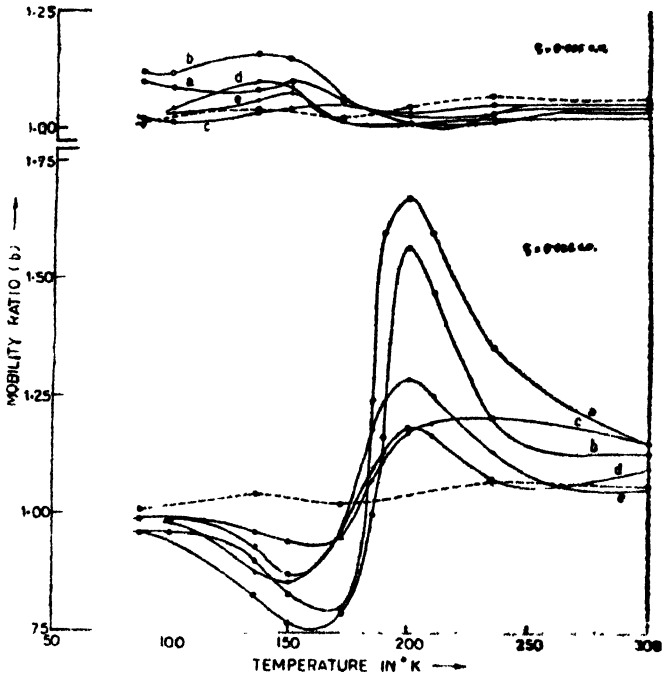

Fig. 2. Temperature variation of b, the ratio of mobilities of electrons and holes for $\zeta = 0.005$ ev. and $\zeta = 0.022$ e.v.

a = crystal 2(a) ... temperature gradient along basal plane.
b = crystal 3(a) ... -do-
c = crystal X_1 ... -do-
d = crystal 1(c) ... temperature gradient along c-axis.

The observed absence of anisotropy in S, however, is not at all improbable, for though the individual mobilities may be different in different directions, their ratios in these directions may remain the same (vide equation above).

It is to be noted here that, we could not properly explain the temperature variation of t.e.p. For this purpose we are now engaged in ascertaining the effect of crystalline defects, which are invariably present in graphite, on the t.e.p. and its temperature variation as also in extending these observations to still lower temperature (20°K) and in studying the effect of magnetic field on t.e.p. The results of these observations will be published soon.

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